

Exciton localization in semimagnetic semiconductors probed by magnetic polarons

K. V. Kavokin and I. A. Merkulov

A. F. Ioffe Physico-Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

D. R. Yakovlev,* W. Ossau, and G. Landwehr

Physikalisches Institut der Universität Würzburg, D-97074 Würzburg, Germany

(Received 22 February 1999)

Peculiarities of optical spectra in diluted magnetic semiconductors are used to find out the characteristic size of the localized exciton state in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys. Three methods for evaluation of the exciton localization radius are suggested. Two of them are based on spin-related phenomena. The localization radius is calculated from measured magnetic-polaron energies and from the degree of the magnetic-field-induced circular polarization of luminescence. The exciton localization radius is determined for a set of samples with Mn content varying from 0.1 up to 0.4. The results of the two spin-related methods are in good agreement to each other. Their comparison with the results of the third approach based on the exciton linewidth analysis allows the conclusion that both the electron and hole of the exciton are localized within the same volume. This latter method, verified by comparison with two others, is applicable also to a wider class of semiconductor compounds which do not contain magnetic impurities. [S0163-1829(99)08347-2]

I. INTRODUCTION

Band-edge features of low-temperature optical spectra in direct-gap semiconductor compounds, and specifically in II-VI solid solutions, are known to be governed by excitons localized at alloy fluctuations. Localization mechanisms have been widely discussed during last two decades (see Ref. 1 and references therein). The importance of these issues for applications, such as engineering of light-emitting devices, is transparent. Fluctuations have an impact on carrier transport, as well as on oscillator strengths and line broadening of light-emitting states of carriers and excitons. On the other hand, the semiconductor compounds allow us to vary easily the degree of disorder by varying alloy composition. Their experimental studies provide a variety of possibilities for testing theoretical approaches to the quantum mechanics of particles in disordered media.

Information on localized exciton states in semiconductor alloys, obtained by traditional absorption and luminescence spectroscopy, is rather limited. In fact, one can measure density of states, oscillator strengths, and lifetimes. It is often difficult to separate contributions of these parameters into the exciton localization. Some progress has been achieved with time-resolved spectroscopic techniques. Now it is possible to gain some information on hopping energy relaxation, dephasing, and the exciton-exciton interaction.² Nevertheless, new experimental information is still needed, especially if it concerns the parameters of localized states which have not been available for direct measurements yet. In this work, we get such information, namely concerning localization radii of exciton states, using diluted magnetic semiconductor (DMS) materials as a model. Properties of their spin systems are employed for probing localized exciton states, besides all the information is extracted from the spectra of polarized photoluminescence under cw optical excitation.

Optical spectra of diluted-magnetic semiconductors, e.g., $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, demonstrate all the basic features inherent to

II-VI compounds. Additionally, they have peculiarities related to the magnetic component. Optical manifestations of the magnetic properties of DMS are due to the strong exchange interaction that couples charge carriers to spins of magnetic ions. The interaction shows up most brightly if a magnetic field is applied to the crystal. The ionic spins, polarized by the field, produce in turn effective exchange fields affecting carriers, whose spin states are as a result split. The splitting energy reaches several tens of meV (giant Zeeman effect).³ In addition to the mean-field-induced magnetization, the magnetic-moment density of the ion spin system will, according to the fluctuation-dissipation theorem, fluctuate. Due to the finiteness of the interaction volume, localized carriers are exposed to exchange fields of spin fluctuations even at zero external field.⁴ These fields split the localized-exciton level to the energy of the order of a few meV,⁵⁻⁷ and make a great impact on the polarization of photoluminescence in weak external magnetic fields.⁸

Another experimentally observed effect, related to the counter influence of carriers onto the spin system, is the formation of magnetic polarons, which are small regions where magnetization is higher than one of the spin fluctuation.⁹ The excess magnetization is induced and maintained by the exchange field of a localized carrier. The carrier reduces its energy at the expense of the interaction with ionic spins by the value varying from units up to tens of meV. This value is usually referred to as the magnetic-polaron energy E_p . It can be determined experimentally from the Stokes shift of the photoluminescence line measured under selective excitation in the tail of localized exciton states.^{9,10}

The giant Zeeman effect shows up in a similar way for free carriers and excitons as well as for localized ones. Therefore, it gives no information on their localization. On the contrary, the splitting of carrier-spin levels by fluctuating fields and the formation of magnetic polarons are extremely sensitive to localization, and are not observed for free particles. Therefore, these phenomena can be employed to study

characteristics of localized states. Qualitatively, it was demonstrated experimentally in quaternary compounds $\text{Cd}_{1-x-y}\text{Mn}_x\text{Mg}_y\text{Te}$, where adding the nonmagnetic constituent, Mg, which gave rise to the alloy disorder leaving the magnetic properties unchanged, resulted in a strong increase of the magnetic polaron energy.¹¹

Here, we report on a quantitative study of the exciton localization, carried out on a representative set of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ samples with various alloy compositions. Three independent methods are suggested. Two of them are based on spin-related effects, i.e., exploit the semimagnetic properties of the studied materials. In the rest of the paper, we first explain the ideas of all three methods and derive the basic formulas. Then the experimental technique and results will be described. Finally, values of localization radii obtained by all three methods will be compared and discussed.

II. METHODS FOR DETERMINATION OF THE EXCITON LOCALIZATION RADIUS

In this section, we discuss theoretical backgrounds for the suggested methods and derive the basic equations which allow us to extract the value of the localization radius from experimental data. In fact, all the three methods described below give the *localization volume* of the localized particle V defined as $V = (\int \Psi^4(\mathbf{r}) d^3\mathbf{r})^{-1}$, where Ψ is the wave function of the particle normalized by the common condition $\int \Psi^2(\mathbf{r}) d^3\mathbf{r} = 1$. The localization radius is defined as $r_0 = [(3/4\pi)V]^{1/3}$. The question to what specific component constituting the exciton (electron or hole) should the measured volume be related, is of importance here. It is considered in each of the subsequent sections and finally discussed in Sec. IV.

A. From the magnetic polaron energy

The exciton magnetic polaron is known to be formed due to polarization of magnetic ion spins by the exchange field of the hole (in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ holes have four times greater exchange constant than electrons, and therefore play the dominant role in the interaction of excitons with magnetic ions).⁹ The carrier-induced exchange field B_{ex} is proportional to the squared modulus of the hole wave function, and thus is in inverse proportion to the localization volume of the hole, V :

$$B_{\text{ex}}(\mathbf{r}) = \frac{1}{3\mu_B g_{\text{Mn}}} \beta J |\Psi(\mathbf{r})|^2 \approx \frac{1}{3\mu_B g_{\text{Mn}}} \beta J \cdot \frac{1}{V}, \quad (1)$$

where β is the constant of the exchange interaction for the valence band, $J = 3/2$ is the hole spin, μ_B and g_{Mn} being the Bohr magneton and the g factor of a Mn ion, respectively. It follows from these considerations that the polaron energy is

$$E_p \approx V \cdot \chi B_{\text{ex}}^2 = \left(\frac{1}{3} \frac{\beta J}{\mu_B g_{\text{Mn}}} \right)^2 \chi \cdot \frac{1}{V} = \left(\frac{1}{3} \frac{\beta J}{\mu_B g_{\text{Mn}}} \right) \chi \cdot B_{\text{ex}}. \quad (2)$$

Thus, having measured the magnetic polaron energy and the magnetic susceptibility χ , one can easily find V .

The susceptibility is conveniently expressed through the parameters of the modified Brillouin function, that describes phenomenologically the magnetic-field dependence of the spin system magnetization in a DMS:¹²

TABLE I. Parameters of the $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys under study used for calculations. Two different values for the magnetic polaron energy E_p are given for alloys with $x < 0.17$ (Ref. 10). The first value corresponds to the MP equilibrium energy determined in time-resolved experiments and the value in brackets is given for cw excitation experiments. These values coincide for $x > 0.17$. For B_{ex} the values determined by means of Eqs. (1), (4), i.e., in the linear approximation for the magnetic susceptibility, are given.

x	T_0 (K)	S_{eff}	θ , (1/T)	E_p (meV)	B_{ex} (T)
0.07	3.1	1.32	4.44		
0.1	4.2	1.08	5.6	10 ^a (1.5)	0.83
0.13	5.4	0.9	5.6	12.9 (8.2)	1.2
0.15	6.0	0.8	5.5	16 (12.8)	1.54
0.16	6.5	0.76	4.26	17 (13)	
0.24	9.5	0.51	2.5	23.3	3.3
0.33	13	0.34	1.39	27	5.2
0.38	15	0.28	1.05	30	7.4

^aExtrapolated value.

$$\chi = \mu_B g_{\text{Mn}} N_0 x S_{\text{eff}} \frac{d}{dB} B_s \left(\frac{s \mu_B g_{\text{Mn}} B}{k_B (T + T_0)} \right)_{B=0} = \frac{s+1}{3} N_0 x S_{\text{eff}} \frac{(\mu_B g_{\text{Mn}})^2}{k_B (T + T_0)}, \quad (3)$$

where N_0 is the number of cation sites in the unit volume, $s = 5/2$ is the spin of a Mn ion, B_s is the Brillouin function for the spin s , k_B is the Boltzmann constant; the effective spin $S_{\text{eff}} < s$, and the effective temperature T_0 , are phenomenological parameters that characterize magnetic properties of the material. The values of S_{eff} and T_0 are determined from the excitonic Zeeman splitting and are well known for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys (see, e.g., Ref. 13). Finally, we obtain

$$V = \frac{\left(\frac{1}{3} \frac{\beta J}{\mu_B g_{\text{Mn}}} \right)^2 \chi}{E_p} = N_0^{-1} x S_{\text{eff}} \frac{s+1}{12} \frac{(N_0 \beta)^2}{k_B (T + T_0)} \frac{1}{E_p}, \quad (4)$$

where for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ $N_0 \beta$ equals 880 meV, and $N_0^{-1} = 56 \text{ \AA}^3$.

It is important to note here that the experimentally observed magnetic polaron energy is nothing else than the shift of the lowest spin sublevel of the exciton due to giant Zeeman effect induced by the exchange field. Having this in mind, one can determine a value of the carrier-induced exchange field B_{ex} with the knowledge on the exciton Zeeman splitting and the magnetic polaron energy. For the $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys studied in this paper the values of B_{ex} are collected in Table I and displayed in Fig. 1 as a function of Mn content.

B. From the polarization of luminescence in magnetic fields

Magnetic fields applied to a DMS align the spins of magnetic ions, inducing a strong exchange field that affects carriers. The splitting of carrier levels in strong magnetic fields (at liquid-helium temperatures these are fields $B \geq 1$ T) is observed immediately as the giant Zeeman effect. At lower fields, the splitting exhibits in the occurrence of polarization

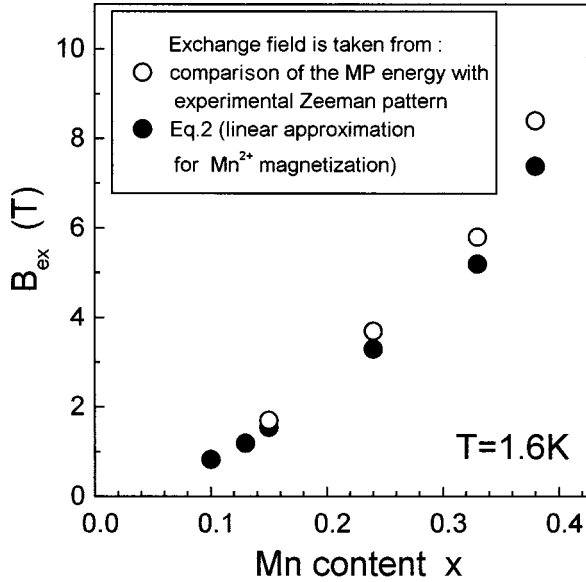


FIG. 1. Values of the exchange field in the exciton magnetic polarons for different $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys.

of the photoluminescence signal. In particular, under observation along the magnetic-field direction (Faraday geometry) the luminescence is circular polarized. This reflects the fact that the spins of the carriers are predominantly aligned parallel to the wave vector of the observed light.

The spin of a trapped carrier is, however, exposed, apart from the mean exchange field, to the exchange field of the local Mn-spin fluctuation. As it was directly shown by the experiment,⁵⁻⁷ the splitting of the hole spin in the fluctuation field is of the order of 2–3 meV. At helium temperatures, this value is much greater than $k_B T \approx 0.1$ meV. For this reason, the spins of localized holes and electrons are completely polarized by local exchange fields which include both the mean component induced by the external field and random fields of spin fluctuations. At these conditions, the polarization of luminescence is governed by the average of the exciton-spin projection onto the direction of light over all the localized states.^{5,8} To calculate the degree of circular polarization, ρ_c , one has to know the mean-squared amplitude of the spin fluctuation. This value can be expressed through the susceptibility χ and the localization volume V by use of the fluctuation-dissipation theorem (FDT). Such a treatment was done in Ref. 8, where the volume V was implicitly expressed via the magnetic-polaron energy along a routine similar to the one described in Sec. II A, and a good agreement with experimental data was achieved.¹⁴ Here, we give a recipe for a reverse procedure, i.e., calculation of V from the measured slope of the dependence of the circular-polarization degree on external magnetic field \mathbf{B} , taken at weak fields:

$$\theta = \left. \frac{d\rho_c(B)}{dB} \right|_{B=0}.$$

Following Ref. 8 we use a Gaussian distribution for the magnetic moment \mathbf{M} of the region where the exciton is localized:

$$\begin{aligned} \varphi(\mathbf{m}, \mathbf{B}) &= \frac{1}{(2\pi)^{3/2}} \exp\left[-\frac{1}{2} \left(\mathbf{m} - \frac{\mathbf{B}}{B_0}\right)^2\right] \\ &= \frac{1}{(2\pi)^{3/2}} \exp\left[-\frac{1}{2} \left(m^2 - 2m \frac{B}{B_0} \cos \gamma + \frac{B^2}{B_0^2}\right)\right], \end{aligned} \quad (5)$$

where γ is the angle between \mathbf{M} and \mathbf{B} ,

$$\begin{aligned} \mathbf{m} &= \frac{\mathbf{M}}{\sqrt{\langle M^2 \rangle}}, \\ B_0 &= \frac{\sqrt{\langle M^2 \rangle}}{d\langle |\mathbf{M}| \rangle / dB} = \frac{\sqrt{(d\langle |\mathbf{M}| \rangle / dB) \cdot k_B T}}{d\langle |\mathbf{M}| \rangle / dB} \\ &= \sqrt{\frac{k_B T}{d\langle |\mathbf{M}| \rangle / dB}} = \sqrt{\frac{k_B T}{\chi V}}. \end{aligned} \quad (6)$$

Deriving the expression for B_0 , we use the well-known result of FDT:

$$\langle M^2 \rangle = \frac{d\langle M \rangle}{dB} \cdot k_B T.$$

The circular polarization degree $\rho_c = (I_+ - I_-)/(I_+ + I_-)$, where I_+ and I_- are intensities of the luminescence components polarized right hand and left hand, respectively. For a heavy hole that has the maximum projection of the spin onto the axis tilted by the angle γ to the direction of observation, $I_+ \propto (1 + \cos \gamma)^2$ and $I_- \propto (1 - \cos \gamma)^2$. By averaging with the distribution function $\varphi(\mathbf{m}, \mathbf{B})$, we get

$$\theta = \sqrt{\frac{2}{\pi}} \frac{1}{B_0} = \sqrt{\frac{2\chi V}{\pi k_B T}}.$$

For a comparison with data of optical experiments it is convenient, as it was done in Sec. II A, to express the susceptibility through the parameters of the modified Brillouin function. This results in the following expression for the localization volume:

$$V = \frac{\pi k_B T \theta^2}{2\chi} = N_0^{-1} \frac{3\pi k_B^2 T (T + T_0)}{2(\mu_B g_{\text{Mn}})^2 x S_{\text{eff}}(s+1)} \theta^2. \quad (7)$$

C. From dependences of the linewidths of exciton absorption and luminescence on the alloy composition

The methods, suggested above for the determination of the characteristic volume of the exciton localization, allow finding V for any specific sample. With the knowledge of data for a set of samples with different alloy compositions, one can realize an independent method of estimating the localization volume, which does not use spin-related effects.

Let us consider the compositional dependence of the width of the long-wavelength wing of the exciton absorption band. This wing is known to be formed by states of excitons localized at alloy fluctuations. As the observed line shape is a result of averaging over a great number of states, one can neglect its fluctuations related to a difference among realizations of the random potential in different samples. This allows us, for theoretical consideration, to substitute a set of

samples with different composition by a single sample where the composition is changed gradually. If the content of, say Mn in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, x increases by a small value δx , the observed change of the long-wavelength wing results from shifts of the levels of localized excitons, induced by an additional random potential $\delta u(\mathbf{r})$. This potential is created by additional atoms of the second component (in our case Mn), randomly distributed over the crystal with the mean concentration $N_0 \delta x$. The mean value of the potential is $\langle u(\mathbf{r}) \rangle = (dE_B/dx) \delta x$, where dE_B/dx is the slope of the dependence of the edge of the corresponding band on x . At low concentrations, $x \ll 1$, additional Mn ions are governed by the Poisson statistics, provided that one considers characteristic volumes much greater than the unit-cell volume. Therefore, we find for the space correlation function of $u(\mathbf{r})$:

$$\langle u(\mathbf{r})u(\mathbf{r}') \rangle = \left(\frac{dE_B}{dx} \right)^2 N_0^{-1} (1-x) \delta x \delta(\mathbf{r}-\mathbf{r}'). \quad (8)$$

The shift of an energy level can be calculated within the first-order perturbation theory:

$$\delta E = \int |\Psi(\mathbf{r})|^2 \delta u(\mathbf{r}) d^3 r, \quad (9)$$

where $\Psi(\mathbf{r})$ is the wave function of the localized particle. It is easy to find that the mean shift of the energy, which equals

$$\langle \delta E \rangle = \int |\Psi(\mathbf{r})|^2 \langle \delta u(\mathbf{r}) \rangle d^3 r = (dE_B/dx) \delta x,$$

is just the change of E_B due to the increase of x . This value is the same for all the states, and therefore does not lead to a change of the linewidth. The line broadening occurs as a result of dispersion of the number of atoms coming into the localization region of the particle. Combining Eqs. (8) and (9), one obtains for the mean-squared energy shift:

$$\begin{aligned} \langle \delta \varepsilon^2 \rangle &= \left\langle \left(\int |\Psi(\mathbf{r})|^2 \delta u(\mathbf{r}) d^3 r \right)^2 \right\rangle \\ &= \left(\frac{dE_B}{dx} \right)^2 N_0^{-1} (1-x) \delta x \int |\Psi|^4 d^3 r \\ &= \delta x \cdot (N_0 V)^{-1} (1-x) \left(\frac{dE_B}{dx} \right)^2. \end{aligned} \quad (10)$$

Equation (10) gives an effective ‘‘inhomogeneous broadening’’ of each localized-exciton line. To calculate the resulting change of the whole spectrum, one has to know its shape. As it is shown in the next section, for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ at energies not too far below the free-exciton level, the Gaussian approximation to the shape of the long-wavelength wing is quite reasonable. By use of Eq. (10), one can find the increment of its variance resulting from the change of composition:

$$\delta(\sigma^2) = \langle \delta \varepsilon^2 \rangle = \delta x (N_0 V)^{-1} (1-x) \left(\frac{dE_B}{dx} \right)^2.$$

From this equation it is easy to derive an expression for the volume in terms of the experimentally measured half-maximum width of the long-wavelength wing of the absorption line, $\Delta E(x)$:

$$V = N_0^{-1} (1-x) 2 \ln 2 \frac{(dE_B/dx)^2}{\frac{d}{dx}(\Delta E^2)}. \quad (11)$$

Equation (11) is immediately applied for evaluation of the volume if the broadening of the exciton line is governed mainly by one type of carriers—electron or hole. Under these conditions, one has to substitute in Eq. (11) composition dependences of the conductance or valence band, dE_C/dx or dE_V/dx respectively, for dE_B/dx . In this case, V has the sense of the characteristic volume occupied by the wave function of either electron or hole in the localized exciton state. If, to the opposite, the radii of electron and hole in the exciton are nearly equal to each other, and dE_C/dx is comparable to dE_V/dx , the best estimation can be obtained by substitution of $dE_g/dx = d(E_C - E_V)/dx$ instead of dE_B/dx in Eq. (11).

III. EXPERIMENTALLY DETERMINED PARAMETERS

In order to check experimentally the consistency of different approaches suggested above, we investigated the photoluminescence (PL) characteristics of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ epilayers grown by molecular-beam epitaxy on (100)-oriented CdTe substrates. Mn content was varied from 0.07 up to 0.38. All experiments were performed with samples immersed in the pumped liquid helium at a temperature of 1.6 K. External magnetic fields generated by a superconducting coil were applied perpendicular to the epilayer surface (i.e., in the Faraday geometry). The photoluminescence was excited by an Ar-ion laser with the photon energy of 2.41 eV. A quarter-wave plate and linear analyzer were used to distinguish right-hand and left-hand polarized luminescence signals. Energies of exciton magnetic polaron (MP) were determined in detail in Ref. 10. In this case a dye laser (Pyridyne, DCM Rodamine) was used for a PL excitation.

It is seen from Eq. (2) that the MP energy is linearly proportional to the magnetic susceptibility χ and the exchange field B_{ex} . As χ is a decreasing function of x for the range $x = 0.15-0.4$,¹⁵ the increasing values of the MP energy (see Table I) should be related to the B_{ex} behavior, namely to the increase of B_{ex} for higher x . Characteristic values of the exchange fields can be evaluated by means of Eq. (2). They are plotted in Fig. 1 for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys of different compositions. B_{ex} is varied from 1 T up to 7 T monotonically, while the manganese content increases from 0.1 up to 0.38. For a comparison, values of B_{ex} obtained by use of experimentally measured Zeeman patterns are also shown. In this case, B_{ex} has been determined as the field that would have induced the Zeeman splitting equal to the doubled MP energy [see discussion of Eqs. (1)–(4)]. Good agreement of these data with values obtained within the linear approximation for the magnetization, used in Eqs. (1) and (2), proves the validity of the approximation throughout our set of samples. Note that the linear approximation is implied also in the calculations of the polarization degree (Sec. II B).

PL excitation (PLE) spectra were measured under dye laser excitation. From the PLE spectra the parameters of the giant Zeeman splitting of the excitonic states T_0 and S_{eff}

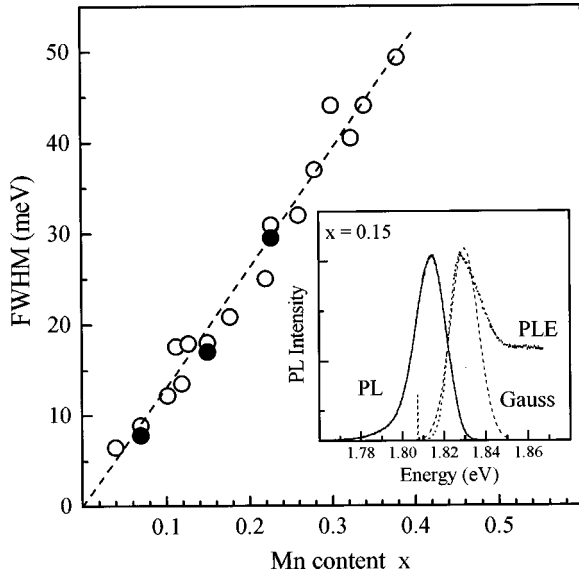


FIG. 2. The full width at a half maximum of the excitonic line in the PL (open symbols) and PLE (closed symbols) spectra of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys. The dashed line is a linear interpolation of the experimental data. The inset shows PL and PLE spectra for a $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$ measured at 1.6 K.

were determined. These parameters for the studied samples are listed in Table I.

The linewidth [i.e., the full width at a half maximum (FWHM)] of excitonic photoluminescence and PLE, the magnetic-field-induced polarization degree $\rho_c(B)$ and energies of exciton magnetic polarons were measured for each sample. These experimental parameters are presented in Figs. 2 and 3 and collected in Table I.

An inset of Fig. 2 depicts typical PL and PLE spectra for a $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$ epilayer, which are dominated by recombination of excitons localized at alloy fluctuations. The PL

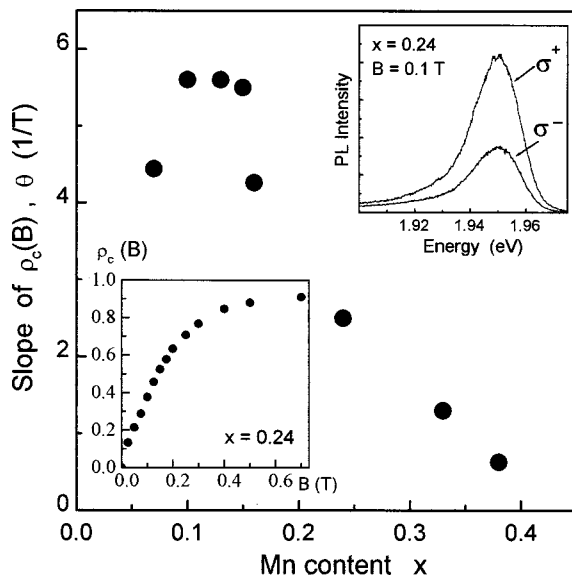


FIG. 3. The slope of the magnetic-field-induced polarization degree of PL as a function of Mn content in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys. Examples of the polarized PL spectra and $\rho_c(B)$ dependence for a $\text{Cd}_{0.76}\text{Mn}_{0.24}\text{Te}$ alloy are shown in the upper and lower insets, respectively, $T = 1.6$ K.

excitation spectrum reflects the density of states for the interband optical transitions. It has a pronounced maximum, which corresponds to the energy of a free exciton. The low-energy wing of the excitonic line is formed by the excitonic states localized at alloy fluctuations. It can be well approximated by a Gauss function shown by a dashed line. The high-energy wing of the PLE excitonic line has a considerable additional contribution due to band-to-band absorption (note that an exciton binding energy of 10 meV is smaller than the discussed linewidths), which causes the asymmetry of the line. We determine the FWHM of excitonic line from the Gauss fit of the low-energy wing of PLE spectra. For the whole set of the studied semiconductor alloys linewidths of the PLE and PL lines give very similar values. Excitonic linewidths of PL and PLE spectra are plotted in Fig. 2 as functions of Mn content. Experimental points can be satisfactorily described by a linear dependence $\text{FWHM}_{\text{PLE}} = 124x$ [meV]. Keeping in mind that PLE spectra reflect absorption spectra and that a FWHM is equal to twice ΔE introduced in Sec. II C, we find that $\Delta E \approx 62x$ [meV]. We will use this dependence below for the evaluation of the exciton localization radius from Eq. (11).

The magnetic-field-induced polarization degree $\rho_c(B)$ presented in the inset of Fig. 3 for a $\text{Cd}_{0.86}\text{Mn}_{0.24}\text{Te}$ alloy shows a linear increase at low fields with saturation at the level of about 1.0 (i.e., 100% polarization). A linear slope of $\rho_c(B)$ at low fields $\theta = d\rho_c/dB|_{B=0}$ is determined by the magnetic susceptibility, which decreases strongly for higher Mn concentrations.¹⁵ The respective decrease of $\theta(x)$ dependence for larger x is clearly observable in Fig. 3.

Energies of exciton magnetic polarons in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys have been reported in Ref. 10 (see Fig. 7 in this reference). They were measured by the method of selective excitation. Localized excitons were excited resonantly at energies below the exciton mobility edge. Under these experimental conditions only the magnetic relaxation via the magnetic polaron formation contributes to the Stokes shift of the PL line away from the excitation energy. The value of this Stokes shift can be associated with the magnetic polaron energy E_p introduced by Eq. (2). For the experiments performed under steady-state excitation conditions, the Stokes shift coincides with the equilibrium polaron energy E_p only if the exciton lifetime τ is longer than the magnetic polaron formation time τ_f , i.e., $\tau_f/\tau < 1$.⁹ It has been established in Ref. 10 that for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys this situation is realized for $x \geq 0.17$. For alloys with $x < 0.17$, E_p can be determined from the saturation level of the polaron shift in time-resolved experiments. In the following, in order to estimate the hole localization volume along Eq. (4), we will take time-resolved data for $x < 0.17$ and steady-state data for $x \geq 0.17$. For $\text{Cd}_{0.9}\text{Mn}_{0.1}\text{Te}$, where the saturation level was not established in the time-resolved experiment, we use an extrapolated value for E_p of 10 meV. The values of the magnetic polaron energies for the studied samples are included in Table I.

IV. COMPARISON OF DIFFERENT METHODS AND DISCUSSION

Composition dependences of the exciton localization radius, determined with different methods, are shown in Fig. 4. The dependences of r_0 on x , determined from the x depen-

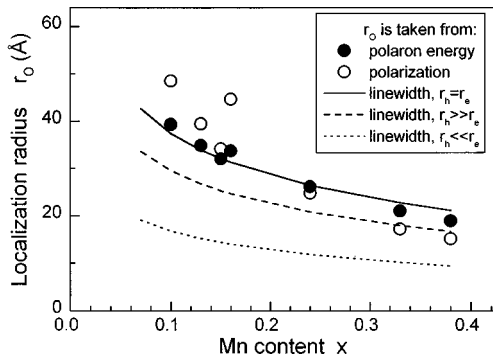


FIG. 4. Comparison of values of the localization radius in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys, determined by different methods suggested in this paper: closed circles are from the MP energy [Sec. II A, Eq. (4)]; open circles are from the polarization of PL [Sec. II B, Eq. (7)]; lines are from linewidths [Sec. II C, Eq. (11)] for different relative values of r_h and r_e .

dence of line widths, are shown by lines for three characteristic cases of the exciton localization: (i) $r_h \gg r_e$ ($dE_B/dx = dE_C/dx$), (ii) $r_h \ll r_e$ ($dE_B/dx = dE_V/dx$), and (iii) $r_h \approx r_e$ ($dE_B/dx = dE_g/dx$). It is important to note here that the methods involving spin-related effects give values of the localization radius of the hole, due to the greater value of the exchange constant in the valence band of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$.

It is seen from Fig. 4 that these methods give dependences $r_h(x)$ which are in a good agreement to each other. A comparison of these results with values obtained from the linewidth analysis allows to conclude about the relation between localization radii of the electron and the hole in the exciton. Actually, $r_0(x)$ calculated assuming $r_h \approx r_e$ proves to be in a very good agreement with the results of spin-related methods. To the contrary, the assumption that the linewidth is governed by the hole, which implies the relation $r_h \ll r_e$, leads to obvious contradiction with the results of spin-related methods, since it gives too small values of r_h . Calculations for the case of the predominant contribution of the electron

(corresponding to $r_h \gg r_e$) give the value of the electron radius, close to one of the hole, thus being in contradiction to the starting premise.

Good coincidence of all methods for the case $r_h \approx r_e$ is not very surprising for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$. In the studied materials 2/3 of the band offset is allotted to the conduction band, but, on the other hand, the electron effective mass is considerably smaller than one of the heavy hole. As a result, a greater amplitude of the localizing random potential for the electron comparative to the hole is compensated by a stronger dependence of the electron kinetic energy on the localization radius. This conclusion is also in line with the results of Ref. 16, where magnetic polaron energy was studied as a function of the hole effective mass under anisotropic strain. It was shown that extreme cases of predominant contributions of either electron or hole in the exciton localization do not provide a good fit to the experimental data.

In conclusion, by use of peculiar properties of diluted magnetic semiconductors, we have attempted an investigation of the exciton localization in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys. The data on localization radii, obtained by three independent methods, are in good agreement to each other. This proves the validity of the starting assumptions. A tentative conclusion can be made that localization radii of the electron and the hole in an exciton are close to each other, which is reasonable for semiconductor alloys with cation substitution. The method to determine the localization radii using the dependences of linewidths on the alloy composition, which is verified in this work by a comparison with spin-related methods, is applicable also to non-magnetic materials.

ACKNOWLEDGMENTS

The authors are thankful to A. A. Klochikhin and A. N. Reznitskii for helpful discussions. This work was supported by the Deutsche Forschungsgemeinschaft through SFB 410, by the Volkswagen Foundation, and by the Russian Foundation for Basic Research Grant No. 96-02-16941.

*Also at A. F. Ioffe Physico-Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia.

¹S. Permogorov and A. Reznitsky, *J. Lumin.* **52**, 201 (1992).

²*Optics of Semiconductor Nanostructures*, edited by F. Henneberger, S. Schmitt-Rink, and E. O. Göbel (Akademie-Verlag, Berlin, 1993).

³J. K. Furdyna, *J. Appl. Phys.* **64**, R29 (1988).

⁴T. Dietl and J. Spalek, *Phys. Rev. Lett.* **48**, 355 (1982).

⁵J. Warnock, R. N. Kershaw, D. Ridgely, K. Dwight, A. Wold, and R. R. Galazka, *J. Lumin.* **34**, 25 (1985); *Solid State Commun.* **54**, 215 (1985).

⁶V. P. Kochereshko, I. A. Merkulov, K. V. Kavokin, D. R. Yakovlev, G. Mackh, W. Ossau, and G. Landwehr, *Proceedings of the International Conference on Semiconductor Heteroepitaxy*, Montpellier, France 1995, edited by B. Gil and R. L. Aulombard (World Scientific, Singapore, 1995), p. 218.

⁷D. R. Yakovlev, V. P. Kochereshko, I. A. Merkulov, G. Mackh, W. Ossau, A. Waag, and G. Landwehr, *Proceedings of the 23rd International Conference Physics of Semiconductors, Berlin, Germany 1996*, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), p. 397.

⁸I. A. Merkulov, D. R. Yakovlev, K. V. Kavokin, G. Mackh, W. Ossau, A. Waag, and G. Landwehr, *Pis'ma Zh. Eksp. Teor. Fiz.* **62**, 313 (1995) [*JETP Lett.* **62**, 335 (1995)].

⁹D. R. Yakovlev and K. V. Kavokin, *Comments Condens. Matter Phys.* **18**, 51 (1996).

¹⁰G. Mackh, W. Ossau, D. R. Yakovlev, A. Waag, G. Landwehr, R. Hellmann, and E. O. Göbel, *Phys. Rev. B* **49**, 10 248 (1994).

¹¹G. Mackh, M. Hilpert, D. R. Yakovlev, W. Ossau, H. Heinke, T. Litz, F. Fischer, A. Waag, G. Landwehr, R. Hellmann, and E. O. Göbel, *Phys. Rev. B* **50**, 14 069 (1994).

¹²J. A. Gaj, R. Planel, and G. Fishman, *Solid State Commun.* **29**, 435 (1979).

¹³W. J. Ossau and B. Kuhn-Heinrich, *Physica B* **184**, 442 (1993).

¹⁴The polarization can be influenced by the magnetic-polaron effect also. Strictly speaking, the equilibrium distribution of spins of localized excitons is determined by alignment of magnetic-polaron magnetic moments in the external field [see Yu. G. Kusrayev and A. V. Koudinov, *Phys. Status Solidi B* **190**, 315 (1995)]. However, as it was shown in Ref. 8, formation of the magnetic polaron drastically slows down the directional relax-

ation of its magnetic moment, so that the equilibrium distribution is not reached during the exciton lifetime. For this reason, the polarization of luminescence is determined by the initial distribution of magnetic moments of spin fluctuations. This conclusion may not be valid for samples with lower x , where polaron formation time is long (see discussion in Sec. III), and in the case when strong localization and/or high magnetic susceptibility leads to saturation of magnetization inside the polaron. One should, therefore, be careful when analyzing data on luminescence polarization. A comparison with estimations of the local-

ization radius obtained from magnetic-polaron energy (see previous subsection) is a valuable *a posteriori* test of validity of the approach.

- ¹⁵S. Oseroff and P. H. Keesom, in *Semiconductors and Semimetals*, edited by J. K. Furdyna and J. Kossut (Academic, London, 1988), Vol. 25, p. 73.
- ¹⁶R. Fiederling, D. R. Yakovlev, W. Ossau, G. Landwehr, I. A. Merkulov, K. V. Kavokin, T. Wojtowicz, M. Kutrovski, K. Grasa, G. Karczewski, and J. Kossut, *Phys. Rev. B* **58**, 4785 (1998).